

Precision frequency measurements with entangled states

Christian F. Roos*

*Institut für Quantenoptik und Quanteninformation, Österreichische Akademie der Wissenschaften,
Technikerstraße 21a, A-6020 Innsbruck, Austria*
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We demonstrate how quantum entanglement can be used for precision frequency measurements with trapped ions. In particular, we show how to suppress linear Zeeman shifts in optical frequency measurements by using maximally entangled states of two ions even if the individual ions do not have any field-independent transition. In addition, this technique allows for an accurate measurement of small external field frequency shifts such as the electric quadrupole shift which are important for ion clock experiments.

Frequency standards based on narrow-linewidth optical transitions in neutral atoms and ions hold great promise as future primary frequency standards [1,2]. In an optical clock, a laser interrogating an atomic transition serves as a local oscillator. The laser frequency is regulated by a servo loop to match the resonance of the atomic transition; a frequency comb divides the optical frequency down to the microwave regime where it can be counted electronically. Laser-cooled single ions with forbidden transitions connecting the ground to a long-lived metastable state have the potential of achieving quantum-limited instabilities of about 10^{-15} [2] for 1s averaging times and fractional frequency uncertainties down to 10^{-18} [3]. To achieve this exquisite level of precision, transitions between atomic levels have to be chosen whose transition frequencies are highly immune to external magnetic and electric fields. Great care has to be taken to determine level shifts caused by unavoidable fields (e. g. black-body radiation) with high precision.

The capability of deterministically entangling quantum systems is a key element in many quantum information and communication protocols. For quantum measurements involving neutral atoms and ions, techniques developed in this context [4] allow for improved parameter estimation in interferometric setups, for efficient quantum state detection [5,6], the measurement of scattering lengths [7] and for beating the shot noise limit. Even prior to the surge of interest in quantum information processing, the use of quantum-mechanical correlations for improving optical clocks was being investigated. It has been shown that N-ion entangled states can lead to a sensitivity not attainable with the same number of uncorrelated atoms [8,9] and first demonstration experiments have been performed [10,11]. This technique is a generalization of Ramsey spectroscopy to entangled states. While the gain in measurement sensitivity due to entanglement between the ions depends on the model under consideration [9,12], it is always smaller than \sqrt{N} and becomes considerable only if large numbers of ions can be reliably prepared in a maximally entangled state.

In this paper, we will discuss a different application of maximally entangled states for high-precision spectroscopy that requires only two ions to be entangled. We will show that certain external-field-dependent level shifts can either be cancelled or precisely measured by preparing the two-ion maximally entangled state in a superposition involving several different internal atomic levels. This leads to two possible applications of entangled states: (1) the measurement of optical frequencies with increased immunity to decoherence caused by fluctuating magnetic fields and (2) the measurement of small frequency shifts caused by external electric and magnetic fields. Recently, it has been shown that entanglement is able to persist for up to 1s, limited only by spontaneous decay of the metastable atomic level [13]. This clearly demonstrates the potential entangled states have for precision measurements.

All ions investigated as potential optical frequency standards [1] have narrow optical transitions connecting the ground state with a metastable excited state that typically has a lifetime of several hundred milliseconds or more. Most of them are either hydrogen-like ions with half-integer nuclear spin such as $^{199}\text{Hg}^+$, $^{171}\text{Yb}^+$, $^{87}\text{Sr}^+$ or have alkali-earth-like spectra, e. g. $^{115}\text{In}^+$ and $^{27}\text{Al}^+$. While these ions have transitions with either zero or very small first-order Zeeman shift, such favorable transitions do not exist in even-isotope hydrogen-like ions such as $^{88}\text{Sr}^+$, $^{40}\text{Ca}^+$, $^{138}\text{Ba}^+$ and $^{172}\text{Yb}^+$. Here, fluctuating magnetic fields may preclude the observation of the natural line width of narrow resonance lines from the $S_{1/2}$ to the $D_{5/2}$ level. Careful magnetic shielding of the apparatus is required to observe narrow lines. From this point of view, even isotopes seem to be much less attractive as frequency standards.

Yet, entangled states make it possible to cancel the first-order Zeeman effect even in those ions that have no first-order-free transitions. The cancellation method is inspired by the technique described in ref. [8] but makes use of the multi-level structure of real atoms. For a single two-level ion with ground state $|g\rangle$ and excited state $|e\rangle$,

separated by the energy $\hbar\omega_0$, the transition frequency can be measured by Ramsey spectroscopy: a laser pulse with frequency ω_L and pulse area $\pi/2$ coherently excites the ion to the state $(|g\rangle + |e\rangle)/\sqrt{2}$. In a reference frame rotating with frequency ω_L this superposition evolves during a period of free evolution of duration τ into the state $(|g\rangle + e^{-i\Delta\tau}|e\rangle)/\sqrt{2}$ where $\Delta = \omega_0 - \omega_L$. A second $\pi/2$ pulse finally maps this state onto an unequal superposition of $|g\rangle$ and $|e\rangle$ so that by measuring the population difference $p_e - p_g = \cos(\Delta\tau)$ information about the difference between atomic and laser frequency is inferred. The Ramsey method is applicable to entangled states in the following sense: The first $\pi/2$ pulse is replaced by a sequence of laser pulses that prepares the two-ion Bell state $(|g\rangle|g\rangle + |e\rangle|e\rangle)/\sqrt{2}$. This state evolves during free precession into $(|g\rangle|g\rangle + e^{-i2\Delta\tau}|e\rangle|e\rangle)/\sqrt{2}$. After applying $\pi/2$ pulses to both ions, a measurement of $p_{ee} + p_{gg} - p_{eg} - p_{ge} = \cos(2\Delta\tau)$ reveals the deviation of the laser frequency from the atomic resonance. Here, p_{ij} denotes the joint probability of detecting ion 1 in state $|i\rangle$ and ion 2 in state $|j\rangle$. By identifying the state $|e\rangle$ ($|g\rangle$) with the eigenstate of the Pauli spin operator σ_z with positive (negative) eigenvalue, this measurement gives the expectation value of the spin correlation $\sigma_z^{(1)}\sigma_z^{(2)}$.

Now, the cancellation of the first order Zeeman effect in even isotopes is achieved by applying the generalized Ramsey method to Bell states of the type

$$|\psi_0\rangle = \frac{1}{\sqrt{2}}(|s_+\rangle|s_-\rangle + e^{i\phi_0}|m'_j\rangle|-m'_j\rangle). \quad (1)$$

Here and throughout the paper, the abbreviations $|m'_j\rangle$ and $|s_\pm\rangle$ denote the states $|D_{5/2}, m'_j\rangle$ and $|S_{1/2}, m_j = \pm 1/2\rangle$, respectively. Fig. 1 shows the electric quadrupole-allowed transitions and the level shifts caused by external fields. By associating each level shifting upwards in a magnetic field with another level shifting downwards by the same amount, the energy of the combined system becomes independent of the magnetic field strength in first order. As a consequence, the corresponding ‘super-transition’ connecting the two-ion ground state to the two-ion excited state is not affected by changing magnetic fields. Its frequency can be probed by applying the measurement scheme described above. For the state $|\psi_0\rangle$ defined in eq. (1), the expectation value yields $\langle\sigma_z^{(1)}\sigma_z^{(2)}\rangle = \cos(\frac{\Delta_+ + \Delta_-}{2}\tau - \phi_0)$, where Δ_\pm is the detuning of the laser frequency from the $|s_\pm\rangle \leftrightarrow |\pm m'_j\rangle$ transition frequencies. An advantage of even isotopes stems from the lack of hyperfine structure which makes the cross-over from the Zeeman to the Paschen-Back regime occur at much higher magnetic fields, resulting in a much smaller second-order Zeeman effect. Therefore, for an accurate calculation of the transition frequency, the Landé factor $g_j(D)$ of the excited level does not need to be known to the same precision as for odd isotopes.

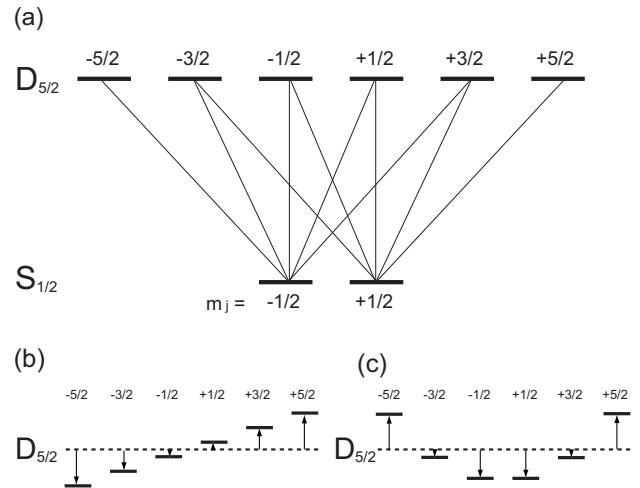


FIG. 1. (a) Zeeman levels of the $S_{1/2}$ and $D_{5/2}$ states and electric quadrupole transitions allowed by the selection rules. (b) In a magnetic field, the energy shift of the Zeeman levels m'_j is $\Delta E_Z = hg_{D_{5/2}}m'_j B$ to first order. (c) Static electric field gradients couple to the quadrupole moment of the $D_{5/2}$ state and shift the levels by an amount $\Delta E_Q \propto 3m'^2_j - j(j+1)$. AC electric fields coupling to the tensor polarizability of the $D_{5/2}$ state lead to level shifts with the same dependence on m'_j . Note that under typical experimental conditions the Zeeman shift is about five orders of magnitude bigger than the quadrupole shift.

Frequency measurements with entangled states require a reliable and deterministic method of producing the entangled initial state. Ion-trap experiments dedicated to quantum information processing [9,14] use strings of ions that are confined in traps providing motional frequencies of a few MHz and giving rise to inter-ion distances of a few μm . After laser cooling one or several normal vibrational modes of the ion crystal close to the ground state, quantum correlations between the internal atomic states are generated by coupling the internal atomic states to their motional degrees of freedom through laser-atom interactions [15,9,16]. This allows for entanglement generation mediated by the motional degrees of freedom. For the generation of ion entanglement, quantum gates or specially tailored series of laser pulses have been used [17,13]. In a non-zero magnetic field, Zeeman transitions can be selectively excited with a narrow-band laser. Generation of the entangled state $|\psi_0\rangle$ is straightforward through the individual addressing of ions with a focused laser beam [14]. Individual addressing by focused laser beams is not, however, absolutely required. Alternatively, since different laser frequencies are needed to excite ions 1 and 2, one could also work with an unfocused beam starting from the state $|s_+\rangle|s_-\rangle$ that could be prepared by an optical pumping beam in a lin \perp lin standing wave. Starting from $|s_+\rangle|s_-\rangle$, the state $|\psi_0\rangle$ defined in eq. (1) could be generated by applying a blue sideband

$\pi/2$ pulse [9] on the $|s_+\rangle \leftrightarrow |m'_j\rangle$ transition followed by a red sideband π pulse on the $|s_-\rangle \leftrightarrow |-m'_j\rangle$ transition.

Are there potential sources of errors that would not occur in frequency measurements with single ions? Slow drifts of control parameters like laser intensity and trap voltage could lead to drifts of the relative phase ϕ_0 of the initial entangled state causing erroneous frequency measurement results. This type of error as well as errors arising from imperfect analysis pulses can be counteracted by setting $e^{i\phi_0} = i$ and by alternating between measurements with long free precession time τ and measurements with τ set to zero. In this way, only changes of the relative phase that occur during the free precession enter the measurement result. The additional measurement will not significantly lengthen the total measurement time since state preparation and detection will typically require a time much shorter than τ .

Also, external field shifts caused by electric fields have to be considered [18]. For two-ion experiments, the use of a linear Paul trap would be mandatory to avoid excess micromotion that would give rise to second-order Doppler shifts and ac-Stark shifts. In contrast to spherical Paul traps, the linear trap's static electric potential $\Phi = Q_{dc}(2z^2 - x^2 - y^2)$, providing the confinement in the axial z direction, produces an electric field gradient $dE_z/dz = -m\omega_z^2/q$ where ω_z is the center-of-mass oscillation frequency of the ion string in the axial direction and m and q are the mass and the charge of the ion, respectively. The interaction of the trap's rotationally symmetric field gradient with the quadrupole moment of the $D_{5/2}$ level shifts the Zeeman levels by an amount [18]

$$\Delta\nu = \frac{dE_z}{dz}\Theta(D, 5/2) \frac{(j(j+1) - 3m_j'^2)(3\cos^2\beta - 1)}{j(2j-1)} \frac{4h}{.} \quad (2)$$

Here, $\Theta(\gamma, j) = \langle \gamma jj | \Theta_0^{(2)} | \gamma jj \rangle$ with $\Theta_0^{(2)} = -e/2(3z^2 - r^2)$ denotes the quadrupole moment and β the angle between the quantization axis and the principal axis of the trap. For a two-ion crystal, the Coulomb potential of the neighboring ion located at a distance $d = (2q^2/(4\pi\epsilon_0 m\omega_z^2))^{1/3}$ adds an electric field gradient to the contribution of the trapping potential so that $(dE_z/dz)_{total} = -2m\omega_z^2/q$ at the equilibrium positions of the ions. However, since any Zeeman state of the $D_{5/2}$ level can be used and $\sum_{m'_j} j(j+1) - 3m_j'^2 = 0$, it is possible to cancel the electric quadrupole shift [19,20] by averaging the measurement results obtained with different m'_j . Yet another effect needs to be considered: the phase evolution of state $|\psi_0\rangle$ is magnetic field-independent only if the field at the positions of ions 1 and 2 is the same. A residual magnetic field gradient in the direction of the ions will change the phase evolution rate. Again, this effect can be averaged out by measuring the phase evolution for state $|\psi_0\rangle$ and for a

state with the roles of ion 1 and ion 2 interchanged [21], i. e. $|\tilde{\psi}_0\rangle = \frac{1}{\sqrt{2}}(|s_-\rangle|s_+\rangle + e^{i\phi_0}|-m'_j\rangle|m'_j\rangle)$.

A second application of entangled states for high-precision spectroscopy does not even require the ultra-stable lasers needed for measurements of optical frequencies. Small level shifts can be precisely determined by preparing Bell states that are immune against phase decoherence caused by a fluctuating laser frequency. This measurement technique should allow for a precise determination of the quadrupole moment of the $D_{5/2}$ level. As can be seen from Fig. 1, the state

$$|\psi_1\rangle = \frac{1}{\sqrt{2}}(|5/2\rangle|-5/2\rangle + |1/2\rangle|-1/2\rangle) \quad (3)$$

is insensitive to the first-order Zeeman shift but evolves in time in the presence of an electric field gradient as $|\psi_1(\tau)\rangle = (|5/2\rangle|-5/2\rangle + e^{i\alpha_1\tau}|1/2\rangle|-1/2\rangle)\sqrt{2}$ where $\alpha_1/(2\pi) = \frac{36}{5}\Delta\nu$ and $\Delta\nu$ is the quadrupole shift experienced by a single ion in state $|5/2\rangle$. The increase in sensitivity as compared to the single-ion state stems from the two-ion entanglement and the fact that Zeeman levels are chosen that are shifted in opposite directions by the field-gradient which itself is two times higher than in the single-ion experiment. If there is a residual magnetic field gradient in the direction of the ion string, then $\alpha_1 = \alpha_{QS} + \alpha_{B'}$ where α_{QS} denotes the contribution of the quadrupole shift and $\alpha_{B'}$ the contribution of the field gradient. The two quantities can be separated by preparing the state $|\psi_2\rangle = (|-5/2\rangle|5/2\rangle + |-1/2\rangle|1/2\rangle)/\sqrt{2}$ where the roles of ion 1 and ion 2 have been interchanged. This state will evolve with phase evolution rate $\alpha_2 = \alpha_{QS} - \alpha_{B'}$ provided that $|\alpha_{B'}| < |\alpha_{QS}|$ so that the quadrupole shift can be measured by taking $\alpha_{QS} = (\alpha_2 + \alpha_1)/2$. Since patch potential effects could produce an additional field gradient, a plot of the quadrupole shift versus ω_z^2 will yield a hyperbola; the quadrupole moment of the $D_{5/2}$ can be determined from the slope of the tangent to $\alpha_{QS}(\omega_z^2)$.

D-state quadrupole moments of $^{199}\text{Hg}^+$, $^{171}\text{Yb}^+$ and $^{88}\text{Sr}^+$ have been recently measured with an accuracy ranging from 3% to 50% [22–24] by detecting Hz-level changes on top of the transition frequencies at $\approx 10^{15}$ Hz. Using entangled states, a relative measurement uncertainty of below 1% is to be expected. For example, in the case of two $^{88}\text{Sr}^+$ ions stored in a linear trap with axial frequency $\omega_z = (2\pi) 1$ MHz, the ions experience a field gradient $dE_z/dz = 72$ V/mm². The phase of $|\Psi_1\rangle$ in eq. (3) would evolve with a frequency of 152 Hz if the direction of the magnetic field was aligned with the trap axis ($\beta = 0$) and using the measured value $\Theta(D, 5/2) = 2.6 ea_0^2$. The entangled state decays within the time $\hat{\tau} = \tau_{D_{5/2}}/2$ where $\tau_{D_{5/2}} \approx 350$ ms is the lifetime of the $D_{5/2}$ level. This sets an upper limit to the maximum useful free precession time τ . Therefore, if the relative phase $\phi_1(\tau) = \alpha_1\tau$ of $|\Psi_1(\tau)\rangle$ after a time $\tau = 150$ ms could be determined with a precision of 0.1

radian, the phase evolution frequency could be measured with an uncertainty of 0.2%. Then the uncertainty in the measured quadrupole moment would probably be dominated by the measurement of the angle β . If the quantization axis could be aligned with the trap's z-axis to within 5° , an uncertainty of below 1% could be achieved.

Employing entangled states for measurements of small energy shifts is not limited to the case of quadrupole shifts: (1) The same measurement principle could also be applied for the determination of the tensor polarizability of the D levels. For this, a transverse static field would have to be applied to shift the ion crystal to a position at which the ions experience a quadratic Stark effect as demonstrated in [24,25]. (2) Another application concerns the measurements of isotope shifts. By loading a crystal consisting of different isotopes, the isotope shift on the quadrupole transitions could be measured with unprecedented precision. (3) Segmented ion traps could be used to separate an ion Bell pair [26]. Then, one ion could be brought close to a surface, thus acting as a miniature sensor of local electromagnetic fields that is read out by the detection of the frequency shift experienced by one ion with respect to the other one. (4) Finally, the search for a possible change of fundamental constants [27] using trapped ions might profit from the use of Bell states. For two ions with similar transition frequencies ν_1, ν_2 but a different dependence on the fundamental constant in question, the measurement sensitivity could be increased by measuring $|\nu_1 - \nu_2|$ in a Bell state instead of detecting ν_1 and ν_2 separately, provided that $|\nu_1 - \nu_2| \ll \nu_1$. The development of frequency combs makes phase locking of the lasers exciting ion 1 and 2 feasible [28].

In summary, we have shown how Bell pairs of ions can be used in precision spectroscopy for absolute frequency and frequency difference measurements. For atomic clocks, entangled states prepared in decoherence-free subspaces might make frequency measurements more robust against environmental perturbations. The use of entanglement for frequency difference measurements might facilitate the detection of tiny frequency shifts on top of the huge frequencies of optical transitions.

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* Electronic address: Christian.Roos@oeaw.ac.at

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